UCRL-15932
S/C 4126105

THEORETICAL ATOMIC AND MOLECULAR PHOTOPHYSICS

J. H. Eberly

August 1987

Lawrence Livermore National Laboratory

B.W. Shore

UCRL--15932

DE87 014249

THEORETICAL ATOMIC AND MOLECULAR PHOTOPHYSICS

J.H. Eberly

Department of Physics and Astronomy
University of Rochester
Rochester, New York 14627

Abstract: We report the results of a theoretical study of long-path stimulated Raman scattering and amplified spontaneous emission in an idealized atomic absorber, under coherent transient conditions and allowing for collisional and Doppler broadening effects.

Final Report of activities carried out under sub-contract 4126105, Lawrence Livermore National Laboratory, 1 Oct. 1983 - 30 Sept. 1986

1. Background

Light travelling through atomic or molecular vapor is absorbed more or less quickly depending on whether the light frequency is close or not to a ground state transition frequency.[1] This is also true for multiple-photon absorptions, and when several light beams of appropriately adjusted frequencies are transmitted together through a vapor cell, the atoms or molecules of the vapor can be excited far from the ground state in a sequence of transitions beginning at the ground state[2] (see Fig. 1a).

For efficient utilization of the photons in such an excitation it is important to work with light beams of sufficient intensity, near to or above saturation level, and to use a cell with adequate absorption path. It is also possible, because of the relatively greater efficiency of generating a given range of frequencies of light, that the first or second transition is arranged to be a true two-photon transition with a far-off-resonant one-photon intermediate state (see Fig. 1b). Under these circumstances secondary atomic and optical processes can occur, such as parasitic amplification of fluorescence, Raman scattering, etc., to an unrelated lower level with the same parity as the initial level (see Fig. 1c). These processes are nuisances because they divert the available light energy from its intended absorption channels.

In the work reported here a study was made of this kind of long-path amplified fluorescence and Raman scattering. Ordinary gain theory has not permitted these parasitic processes to be well understood theoretically up to the present time. We have studied them with enough detail to include the case that they interfere with each other. Interference can occur because both processes depend on the same dipole moments, particularly the moments associated with the far-off-resonance intermediate level labelled 2 here, which is weakly accessible via one pump photon assisted by collisional energy transfer. In view of the difficulty in dealing with long-path propagation of three light pulses simultaneously (laser pump, amplified fluorescence, stimulated Raman light), especially under conditions where saturation of the latter two exists, we were not able to take into account the two-photon on-resonant interaction of the (presumed) second excited state.

Experimental data is invaluable in judging a partially idealized theory. Data for the situation we studied has existed[3] for almost a decade. It is still partly unpublished[4] due to the lack of an adequately detailed theory. However, we have been able to use it for comparison with the predictions that follow from our studies, and find remarkably good agreement. On this basis it appears that our approach may be applicable in realistic situations despite its simplicity (it does not include the level degeneracies that are present in the experimental atoms, for example).

2. Relation to thesis research of B.J. Herman

The study undertaken of the effects described above comprised the Ph.D. thesis research of B.J. Herman in the Department of Physics and Astronomy, University of Rochester. This research was supervised by J.H. Eberly, Professor of Physics and of Optics, University of Rochester. A copy of the thesis is attached to this report as Appendix A. For economy of writing, the developments described in the thesis will not be separately described in detail in this report but will be incorporated through specific references to sections and equations of the thesis, using the initials BJH.

3. Description of Method

Our method is an extension of the method used for analyzing the propagation of multiple simultaneous coherent pulses developed by Konopnicki and Eberly.[5] The foundation of the method is the coupling of Maxwell and Bloch equations[6], as appropriate to the atomic system under consideration (BJH, Chap. II). The novelties, and therefore difficulties, presented by the case at hand are significant, however.

In the present situation there are three simultaneous pulses instead of two, and of the three, two of them interact with exactly the same atomic dipole moment, but with different frequencies. The Raman and fluorescence pulses start, in principle, from spontaneous noise. The initial pulses have been modeled instead with coherent input "seed" pulses. The results have been checked to be independent of the duration and frequency content of the seed pulses over a wide range.

In order to amplify the Raman and fluorescence pulses the dipole moment associated with the 2-3 transition (see Fig. 1c) must oscillate at two frequencies, and so must simultaneously be associated with two separate slowly-varying amplitudes in the rotating wave approximation. The consequence is that the three-level system has effectively not $3x3 = 9$ but 13 significant density matrix elements or Bloch variables (BJH, Sec. 2.5).

Some reduction of this complication can be achieved whenever the collisional effects are not large enough to put more than about 10 per cent of the total population in the off-resonant intermediate level. In this case a partial adiabatic elimination procedure is valid, and we have used it (BJH, Sec. 2.7).

A full treatment of three-pulse propagation and interaction, even with adiabatic elimination in an idealized absorbing atom, must be mostly numerical if saturation is to be included. However, some insights are possible from a futher simplified analytic treatment of the pulse-atom interaction. This is described in BJH, Ch. III, where it is shown that the initial growth of fluorescence can dominate the Raman light when the rate of collisional transfer of population is much larger than the inverse pump pulse width. However, the degree of amplification of fluorescence is determined by the amount of population transferred by the pump-collision process and this is always a small fraction of the ground state population. Since the Raman light feeds off of the latter, it eventually overtakes the fluorescence, even though it results from an inherently weaker two-photon process.

4. Description of Numerical Results

In Figure 2a we show some of the experimental results of Raymer and Carlsten[7], plotting fluorescence and Raman intensities as a function of laser pump intensity. In Fig. 2b we show the results of the theory as well as the experimental data. The agreement can be regarded as only qualitative because of the systematic uncertainties associated with the data as well as because of the simplicity of the theoretical model. Nevertheless the alignment, relative slopes and critical saturation turnover are modelled correctly here for the first time.

In addition to the results shown in Fig. 2b, the theory developed in this study predicts that coherence effects are possible in both the fluorescence and the stimulated Raman light. These are shown in the sequence of graphs contained in Figs. 3 and 4. Perhaps particularly interesting are the development of quasi-stable peaks of area 2π in the amplified fluorescence (Fig. 3) and then after longer path propagation, coherent two-photon self modulation of the pump and Raman light (Fig. 4).

The coherence effects suggested in Figs. 3 and 4 are the subject of a separate paper[8] by B.J. Herman and J.H. Eberly, which is attached as Appendix B. A description of these effects is given there and will not be repeated here.

5. Conclusions

In conclusion we can say that a specific method of theoretical analysis has been developed, and developed to the point of application to existing experimental data. The method of analysis is new in several respects:

a) three separate propagating pulses are treated dynamically at the same time, and allowance is made for pump depletion as well as fluorescence and Raman amplification;

b) the equations of the theory permit coherent interactions, and sufficiently high intensities to be compatible with observed saturation effects;

c) the density matrix equations (or generalized Bloch equations) used in the theory have multiple dipole oscillation frequencies, so that the number of independent slowly varying dipole amplitudes is 4 greater than the number suggested by previous treatments of adiabatically relaxed three-level systems.

The degree of agreement between the theoretical predictions obtained here and the corresponding experimental data is good. This suggests that the nonlinear coupling contained in the coupled Maxwell-Bloch framework is adequate for a good description of fluorescence amplification in company with stimulated Raman scattering, in the presence of collisions, even if the details of level degeneracies and line broadening are not fully included. The theory appears to be sturdy enough to deal with these effects in much greater detail, and to permit application to more elaborate absorption-amplifiation scenarios without complications other than increased computer time. Two elaborations that would be of fundamental interest would be: (1) the inclusion of statistical effects related to the true spontaneous initiation of the pulses [avoiding the need to use artificial "seed" pulses]; and (2) the inclusion of a higher two-photon-resonant level.

6. References

1. L. Allen and J.H. Eberly, Optical Resonance and Two-Level Atoms (Wiley, New York, 1975), Chap. 4

2. See, for example, W. de Ruiter, "Laser Separation of Isotopes", Endeavour New. Ser. (GB) 8, 128 (1984), and B.W. Shore, Theory of Coherent Atomic Excitation (Wiley, New York, to be published).

3. M.G. Raymer and J.L. Carlsten, Phys. Rev. Lett. 39, 1326 (1977); J.L. Carlsten and M.G. Raymer, in Laser Spectroscopy III, ed. by J.L. Hall and J.L. Carlsten (Springer-Verlag, New York, 1977, p. 205)

4. M.G. Raymer, Ph.D. Thesis, University of Colorado, 1979.

5. M.J. Konopnicki and J.H. Eberly, Phys. Rev. A 24, 2567 (1981)

6. See Ref. 1, for example.

7. See Refs. 3 and 4.

8. B.J. Herman and J.H. Eberly, submitted to Optics Comm.

7. Figure Captions

Fig. 1. Schematic diagrams of atomic energy levels participating in multiple-photon excitation leading to ionization. (a) Two-photon excitation leads to ionization through a resonant intermediate state; (b) Two-photon excitation leads to ionization without a resonant intermediate state; (c) Parasitic de-excitation processes (Raman scattering and fluorescence) are shown robbing excitation probability from an off-resonant intermediate level.

Fig. 2. Output photons as a function of normalized input (pump) photons. (a) Experimental data from ref. 7. Note that the three stimulated Raman scattering points are circled for clarity. The other points represent amplifed fluorescence. (b) Comparison of experiment and theory, as descripted in BJH. Due to the theoretical use of coherent probe input fields, the lack of coincidence in the low-intensity portions of the experimental and theoretical curves is not significant. Note the relatively accurate modeling of the portions of the data indicating saturation.

Fig. 3. Theoretical curves from ref. 8, plotted at successively deeper propagation distance in the amplifying medium. The rapid onset of amplified fluorescence is evident, as is the subsequent rise of stimulated Raman intensity.

Fig. 4. As in Fig. 3, showing deeper propagation, and coherent Raman transients reminiscent of two-photon self-induced transparency.

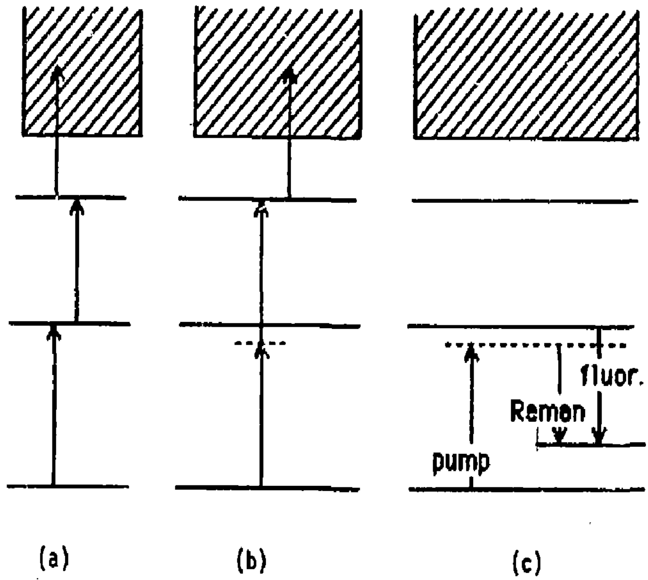
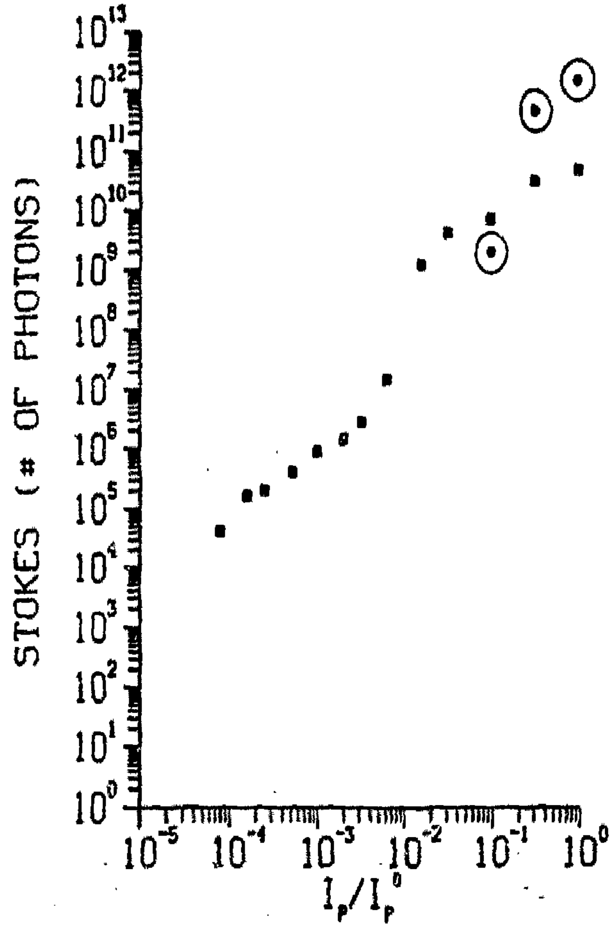
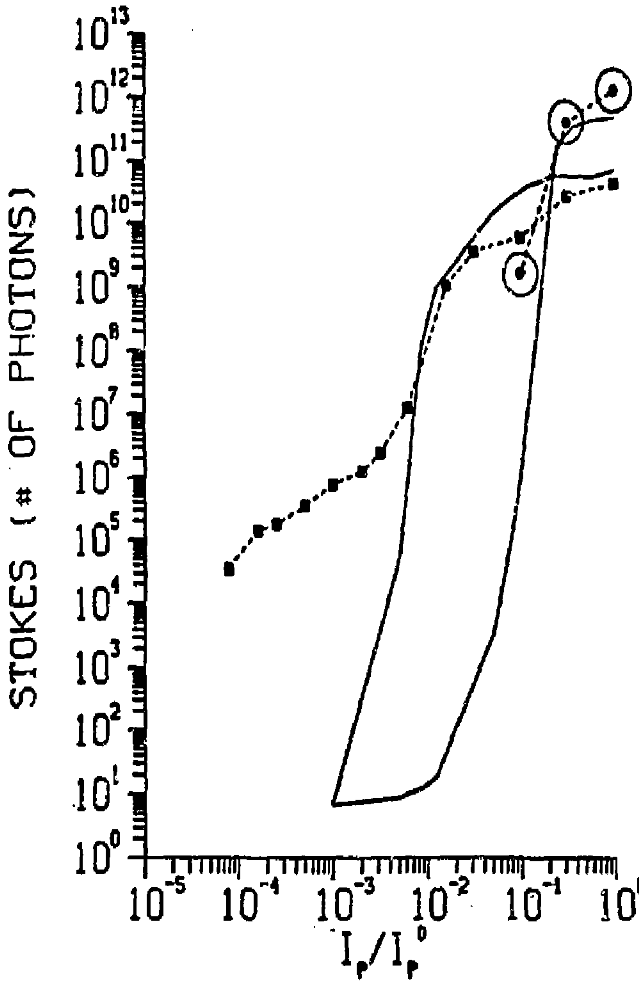


Fig. 1


STOKES (# OF PHOTONS)
10^0
10^1
10^2
10^3
10^4
10^5
10^6
10^7
10^8
10^9
10^10
10^11
10^12
10^13
10^-5
10^-4
10^-3
10^-2
10^-1
10^0
I_p/I_p^0

<u>Fig. 2 (a)</u>



STOKES (# OF PHOTONS)
10^0 10^1 10^2 10^3 10^4 10^5 10^6 10^7 10^8 10^9 10^{10} 10^{11} 10^{12} 10^{13}
10^{-5} 10^{-4} 10^{-3} 10^{-2} 10^{-1} 10^0
I_p/I_p^0

Fig. 2 (b)

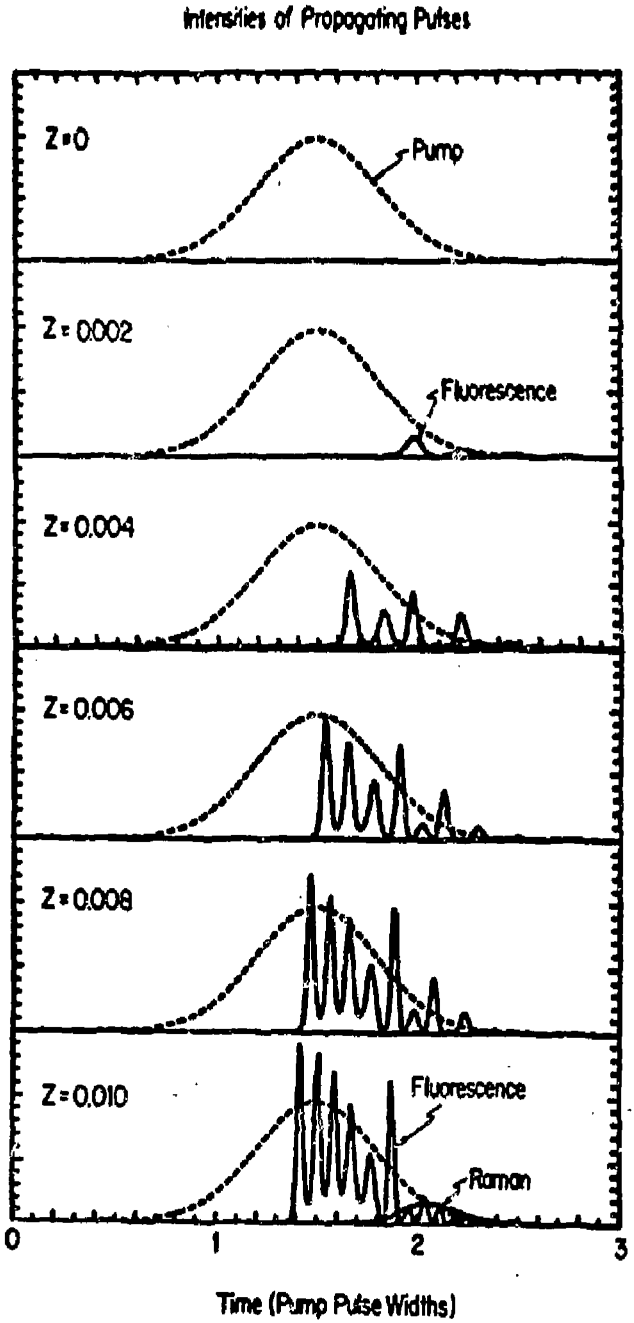
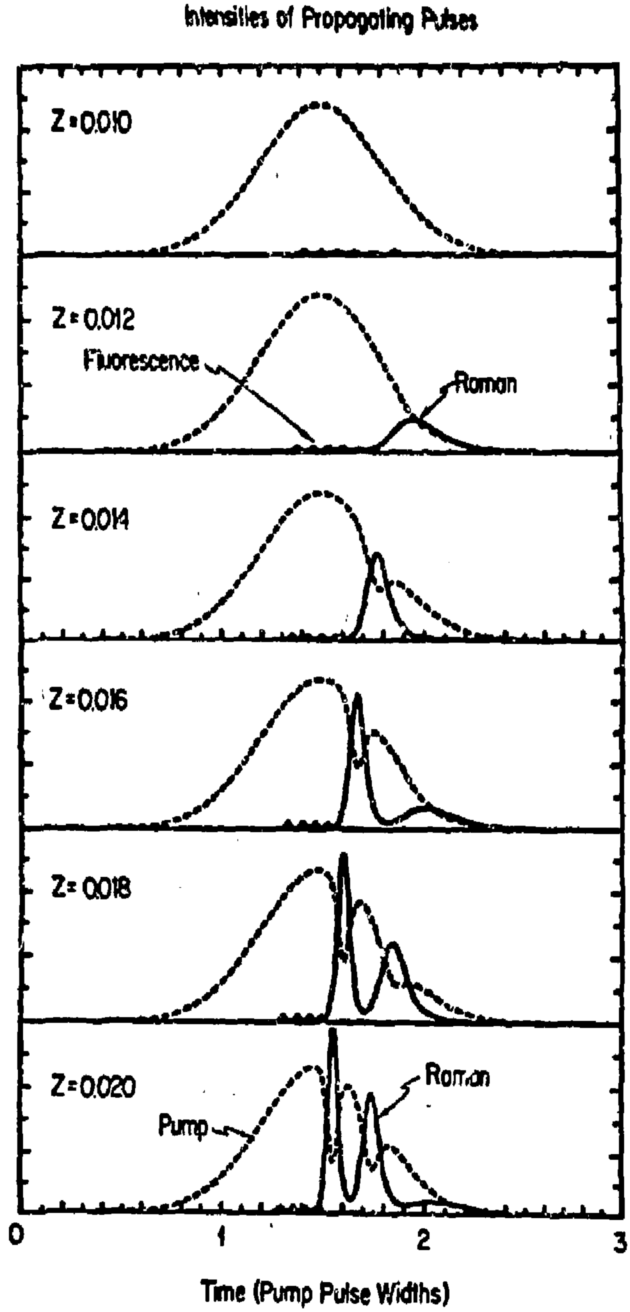

Intensities of Propagating Pulses
Z=0
Pump
Z=0.002
Fluorescence
Z=0.004
Z=0.006
Z=0.008
Z=0.010
Fluorescence
Raman
0
1
2
3
Time (Pump Pulse Widths)

Fig. 3



Intensities of Propagating Pulses
Z=0.010
Z=0.012
Fluorescence
Raman
Z=0.014
Z=0.016
Z=0.018
Z=0.020
Pump
Raman
0
1
2
3
Time (Pump Pulse Widths)

Fig. 4